JOINING OF CERAMICS FOR HIGH TEMPERATURE APPLICATIONS

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Martti Vilpas

Translation of "Korkeissa lampotiloissa kaytettavien keraamisten materiaalien liittaminen". Technical Research Center of Finland, Espoo, Report VTT-TIED-481. August, 1985, pp. 1-37.

(NASA-TT-20030) JOINING OF CERAMICS FOR HIGH TEMPERATURE APPLICATIONS (NASA) 28 p
Avail: NTIS HC A03/MF A01 CSCL 11B

N87-29678

Unclas
G5/27 0100021

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
WASHINGTON, D.C. 20546 October 1987

STANDARD TITLE PAGE

1. Report No. TT-20030	2. Government Accession No.	3. Recipient's Catalog No.
4. Title and Subtitle JOINING OF CERAMICS FOR HIGH TEMPERATURE APPLICATIONS		5. Report Date October 1987 6. Performing Organization Code
7. Author(s) Martti Vilpas		8. Performing Organization Report No. 10. Work Unit No.
9. Performing Organization Name and Address SCITRAN Box 5456 Santa Barbara, CA 93108		11. Contract or Grant No. NASw-4004 13. Type of Report and Period Covered Translation
12. Sponsoring Agency Name and Address National Aeronautics and Space Administration Washington, D.C. 20546		14. Sponsoring Agency Code

15. Supplementary Notes

Translation of "Korkeissa lampotiloissa kaytettavien keraamisten materiaalien liittaminen". Technical Research Center of Finland, Espoo, Report VTT-TIED-481. August, 1985, pp. 1-37.

16. Abstract

The report is a summary of literature survey to the methods for joining ceramics to ceramics or ceramics to metals for high temperature applications. Also mechanical properties and potential applications of the joints are considered.
The joining of ceramics is usually carried out by brazing or diffusion bonding. Especially the latter has been found useful and increased the applications of joined ceramics. Recently also the possibilities to use electron beam and laser beam welding for joining of ceramics have been investigated. The joining of ceramics has found numerous applications, typical for high operating temperatures being different sensors and thermocouples.

17. Key Words (Selected by Author(s))	18. Distribution Statement
	Unclassified and Unlimited

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of Pages	22. Price
Unclassified	Unclassified	25	

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Foreword

This report contains the project of the "New Materials to be Used at High Temperatures", begun in 1984 in the State Technical Research Center (VTT). The project is part of the VTT machine shop industry production technology research program. The objective of the project is to assure the effectiveness of materials to be used at high temperatures, and their safety, by choosing materials as fireproof as possible. The project is being conducted by a search of the literature and by laboratory and field testing in collaboration with the VTT Metallurgical Laboratory, Metals Laboratory and Domestic Fuel Laboratory.

The author would like to thank Associate Professor Kazutoshi Nishimoto of Osaka University for a survey made for the VTT on ceramic bonding technology.

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1. Introduction

Metallic building materials intended for high temperatures (such as tempered steels and fireproof superalloys based on Ni and Co) do not sufficiently withstand mechanical and chemical stresses associated with heat energy, production and other processes (abrasion, oxidation, heat corrosion) under all circumstances. This has excluded them from use in heat energy machinery and power plants, and has increased their maintenance costs. These difficulties have been reduced by means of ceramic building materials. Ceramics are used for high operating temperature ceramics, e.g., aluminum oxide (Al_2O_3), zirconium oxide (ZrO_2), silicon carbide (SiC), silicon nitride (Si_3N_4), and so-called "sialon" (Si-Al-O-N) compounds (Juvonen, 1984). Accordingly, the more solutions reached regarding problems associated with their toughness and bonding technology, the more general the use of these materials for high temperature structures becomes.

The purpose of this study of the literature is to present a summary of the procedures by which ceramic components, intended for high temperature applications, can be joined to other ceramics or to metallic structures. The characteristics and applications of the joints are also treated. Composite structures and ceramic coatings are not considered in this respect.

2. Ceramic Component Bonding Procedures

The bonding of ceramic materials to other ceramic or metallic structural parts has been very problematic. The reason for this is the high sensitivity of ceramic materials to the concentration of local strains. This easily leads to heat stresses causing bonds to crack, especially when heat expansion coefficients in the bonding ceramic and metallic components (Table 1) clearly deviate from one another. The use of ceramic bonds is therefore limited to places where other structural solutions have proved to be unsuitable. Thus bonding has generally been accomplished by brazing. However, recent developments in diffusion welding technology have made cheaper and more economic bonding of ceramic components possible (Gruenling, 1973; Bailey and Borbidge, 1980).

2.1 Brazing

Ceramic component brazing procedures can be divided into three groups (Gruenling, 1973; Christensen and Sheward, 1982):

--Coating ceramic surfaces with metal and brazing with a metallic binding agent

--Brazing with a ceramic binder; and

--Brazing with a metallic binding agent.

2.1.1 Procedure of Metallic Coating on Ceramic Surfaces

A basic problem in brazing ceramics has been the question of poor ceramic surface contact in metallic brazing. Because of this, the brazing of ceramic components has traditionally presupposed a metal-coated ceramic surface, produced by either electrolysis or heat treatment. Fireproof materials, such as W, Mo and Nb, have been used as surface coating materials (Gruenling, 1973; Elsaner, 1975).

The electrolytic lining assumes that the ceramic component is made an electrical conductor by precipitation on the bonding surface of a thin metallic film, e.g., with a gas phase liner. Any metallic coating is able to satisfy this procedure.

Table 1. Physical and mechanical properties of ceramics and metals to be used at high application temperatures (Juvonen, 1984; Anon., 1982 a, b; Nail, 1984).

	1	2	3	4	5	6
7						
Al_2O_3	3950	8,5	≥ 1100	T 350 - 450 P 3800 - 3900	T 220 (1600°C) 280 (1300°C) 320 (1400°C)	1700 - 1900
ZrO_2, 8 stabilized	5900	9	1100 - 1500	T 250 - 300 P 3000 - 3200		2200 - 2600
ZrO_2, 9 mixtain stabilized	5900	7,5	≥ 1100	T 300 - 700 P 4000 - 6000	T 400 (600°C) 220 (1000°C) 225 (1200°C)	
SiC, B- ja C-sementoim 10	3100	3,5	≥ 2500	T 400 - 500 P 3000 - 3500	silliytab 11 lujuutuman n. 1400°C:een 12	1400 - 1600
Si_3N_4, 13 sintrattu puristaen	3300	2,9	≥ 2000	T 700 - 1000 P 6000 - 8000	T 400 (1000°C) 400 (1200°C 300 (1400°C)	1200 - 1400
Si_3N_4, 14 sintrattu upotuksen	2600	3,3	1500 - 1700	T 150 - 200 P 1000 - 2500	silliytab 11 lujuutuman n. 1200°C:een 15	
16 Välihuone-metallerja [2]						
Nb	8400	6,9	115	V 200 - 450	-	-
Pt	21450	9,1	40 - 105	V 125 - 205	-	-
Au	19300	14,2	25 - 65	V 120 - 220	-	-
Mo	9500	13,3 (20 - 100°C)	90	V 340	-	-
Ta	6550	6,5	110 - 180	V 240 - 440	-	-
17 Tulenkestävät metalliseokset						
18 Austeniittiset ruostumattomat teräkset	7900 - 8000	15 - 19	200 (AISI 304)	V 450 - 650 (AISI 304)	V 450 (500°C) 350 (700°C) 200 (800°C) (AISI 304)	
Ni-pohjaiset 19 superseokset	7720 - 8550	11 - 18	120 - 250	V 750 (Inconel 617)	620 (540°C) 520 (760°C) 170 (982°C) (Inconel 617)	
Co-pohjaiset 19 superseokset	8200 - 9200	16 - 18		V 750 (Star H 554)	350 (650°C) 500 (815°C) (Star H 509)	

20 [1] T = taivutuslujuus, V = vetolujuus, P = puristuslujuus
21 [2] käytettiin yleisesti huoneilmakehässä-lämpötilassa välihuoneenlämpötilaa (ks. kohta: diffusiohitsaus)
22 [3] kuuma kovametallisuutta käytettäessä

Key: 1. density, 2-heat expansion coefficient, 3-hardness, 4-strength, 5-heat resistance, 6-maximum application temperature, 7-ceramics, 8-stabilised, 9-partly stabilized, 10-B and C alloys, 11-maintains strength, 12-to about 1400°C, 13-compressed sintrate, 14-nitrified sintrate, 15-to about 1200°C, 16-interposed metals, 17-fireproof metallic alloys, 18-austenitic stainless steels, 19-based superalloys, 20-[1]T = bending strength, V = tensile strength, P = compressive

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Table 1 key continued: strength, 21-2) used mainly as interface plate for ceramic-ceramic bonds (e.g., spot: diffusion welding) 22-3) refers to unloaded application points.

The tensile strength of the electrolytic, metallic-coated brazing bonds is set in the interval from 40 to 100 N/mm^2 (+20°C). Appropriate linings and heat-dependent brazing additives can be used to achieve application temperatures of 800-1000°C for joints (Gruenling, 1973).

Metallic coatings can be applied in heat treatment by using, e.g., Mo or W solders with which oxide binding agents are mixed (Al_2O_3, BaO, CaO, MgO and/or Y_2O_3). The solder spread on the ceramic surface is heated above the fusion temperature of the binding agent on the firm ceramic surface (e.g., Al_2O_3 + CaO or Al_2O_3 + Y_2O_3 heated to 1700-1900°C). The purpose of the oxide binding agent is to form an effective ceramic surface based on a strong chemical bond.

Table 2 presents examples of brazing bond strength and application temperatures in using metallic linings of W-2%Y_2O_3 achieved through heat treatment. The maximum bond strength is 85-155 N/mm^2 as a function of the solder. The highest strength of 180 N/mm^2 is achieved with W-5%Y_2O_3 linings. Application temperatures are even possible above 1000°C, as a function of the heat resistance of the solder and the fusion point of the oxide binder.

The welding and the double-phase nature restrict the use of a prebrazing procedure. For this reason metallic coatings are used today only in special cases, because single-phase brazing procedures and additives are tangibly faster.

2,1.2 Ceramic Solders

Metallic coatings for ceramic components are not necessary when an oxide alloy is chosen as the solder binding agent (usually Al_2O_3, MnO and SiO alloys). These so-called ceramic solders have been used to produce ceramic-ceramic bonds and bonds between ceramic components and fireproof metals, such as W and Ta.

Table 2. The tensile strength of metal-coated, brazed ceramic bonds lined with W-$25Y_2O_3$ with various additives and parameters (vacuum brazing $\sim 10^{-3}$ Pa) (Gruenling, 1973).

1 Juote	Juottoparametrit		Juotesaumakoon vetolujuus +20 °C, N/mm²		Lujuuteen ja käyttöihanteisiin liittyviä huomautuksia
	lämpötila (°C) 5	aika (s) 6	min - max	keskim. 7	
Cu-28%	1130	180	125 - 155	140	T(käytt.) < 900 °C. Hyvä lujuus (+20°C) saavutetaan, koska juote ei seostu metalliolintikerrokseen. Vetokokeen murtuma keramiikassa.
Pd	1370 1570	20 90	65 - 125 80 - 110	100 95	Korkea T(käytt.) < 1200 °C. Juote seostuu metalliolintikerrokseen, josta seuraa alhaisempi lujuus. Vetokokeen murtuma rajapinnalla: keramiikka-metalliolinti.
V-30Nb-37%	1805	60	80 - 85	82	Korkea T(käytt.) < 1200°C. Liitoksen alhaisempi lujuus johtuu juottoon seostumiseen metalliolintikerrokseen. Liitoksen kestävyys sykliseen kuumennukseen todettu hyväksi (100 = 100-1200°C).

Key: 1-solder, 2-brazing parameters, 3-brazed bond tensile strength, 4-comments on strength and application temperatures, 5-temperature, 6-time, 7-average, 8-T (application < 900°C. Good strength (+20°C) achieved when solder is not alloyed in metallic layer. Fracture of ceramics in tensile test. 9-High T (application) < 1200°C. Solder alloyed in metallic layer from which lowered strength comes. Tensile test fracture at boundary surface: ceramic-metal. 10-High T (application) < 1200°C). Lower strength of bond leads to alloying of solder in metallic layer. Durability of bond in cyclical heating has turned out well (100 = 100-1200°C).

Brazing is conducted in a furnace at a temperature at which the oxide alloy placed between the surfaces melts and wets these bonding surfaces (Al_2O_3, MnO and SiO alloys: $T \gtrsim 1140$°C. Bonding results from the diffusion and the chemical reactions occurring between the surfaces being bonded.

Generally the use of chemical solders comes into consideration only if the heat expansion coefficients of the components to be joined are close to one another (Table 1). Generally the maximum application temperature of the bonds remains below

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750°C. However, with binary, tertiary and quaternary components of Al_2O_3-CaO (-Mg, -SiO), solders attain application temperatures above 1700°C. The maximum bonding strength of bonds brazed with ceramic additives are recorded between 170 to 185 N/mm^2 (Gruenling, 1973).

The adherence of the ceramic bonds to the metallic surfaces can be improved by the so-called Cermeting process, in which an oxide film is sintered onto the fireproof surface of the metallic component before the brazing (e.g., an Mo-Y_2O_3-Al_2O_3 film, sintered at 1900°C, onto an Mo surface). Then a eutectic Y_2O_3-Al_2O_3 alloy (fusion temperature T_m = 1760°C) can be used as a brazing additive. This is forced into the Cermet surface pores and achieves a strong adhesion between the components. However, the procedure has the drawback of being slow and having two phases.

2.1.3 Metallic Solders

The simplest way to prepare a brazed bond between ceramic components is to use "active" metallic additives, which wet the bond surfaces without any need for pre-treatment. The basic prerequisite of such solders is their ability to thoroughly wet the ceramic surface. Also typical of the metallic solders is their activity, e.g., their great affinity for oxygen, where the solder reacts with oxidized ceramics, and the oxide film growing on the bond surface achieves good adherence on its ceramic surface. The connection is formed by diffusion between the surfaces and by chemical reactions.

Soldering success assumes the correct choice of parameters (temperature, time), for an oxide layer becoming too thick on a bond surface reduces the bond strength (Gruenling, 1973; Naka et al., 1984a). For this reason easily oxidized metallic bonding agents are usually soldered in a vacuum (10^{-2} - 10^{-3} Pa) or in protective inert gases.

Metallic solders fit for high temperature are, e.g., Ti and Zr alloys, to which various percentages of Cu or Ag are added. Application temperatures of some 800°C are achieved with these solders (Gruenling, 1973; Klaaner, 1975). A very wettable solder of 50% Cu and 50% Ti can also be used for soldering, Figure 1. This type of solder includes the use of Al_2O_3 and Fe-Ni-Co alloys between bonds, where a cutting strength of 165 N/mm^2 is attained. The maximum application temperature of Ko bonds is limited to the 600-700°C range, where the cutting strength of the bond has been observed to drop rapidly from a level of 125 N/mm^2 to a value of 50 N/mm^2 (Naka et al., 1984a).

Ternary Zr-19%Nb-6%Be, Ti-48%Zr-4%Be and V-30%Nb5%Ti alloys are used in soldering ceramic-metallic bonds intended for high temperature application. Using these additives makes it possible to eliminate the formation of compounds between brittle metals in a bond. These solders are used with good success with W in bonding metal-coated ceramic compounds (Gruenling, 1973).

2.2 Welding

2.2.1 Diffusion Welding

Diffusion welding is promising and is used most widely today for processes bonding ceramic components. The principle of the process is an old one, and has already been applied in so-called machine welding, one of the oldest joining processes. One of the most important areas of diffusion welding today is bonding different kinds of materials.

Principle

Bonding is achieved in diffusion welding by simply pressing dressed and cleaned surfaces to be bonded together and heating them in vacuum and protective gas chambers. Cohesion and creep are achieved by atomic contact between the bond surfaces. The heat action of the atoms makes the diffusion between the bond surfaces possible, and leads to the formation of the diffusion bonds, Figure 2. In this process both metals and ceramics can be joined by chemical reactions, in which

adherence can be based, for example, on oxide growth on the bond surface.

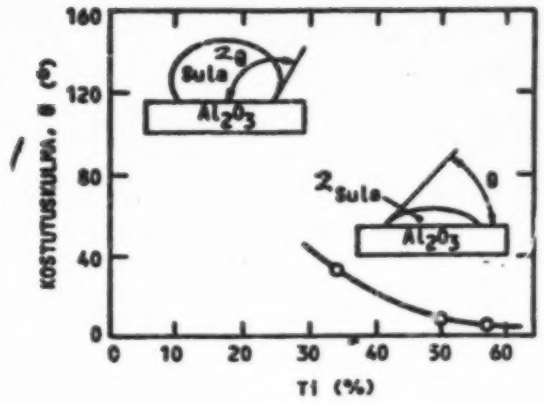

Figure 1. Effect of Ti content on the wetting ability of an Al_2O_3 surface of Cu-Ti solders (1025°C, 30 minutes). (Naka et al., 1984b). Key: 1-wetting front, 2-melt.

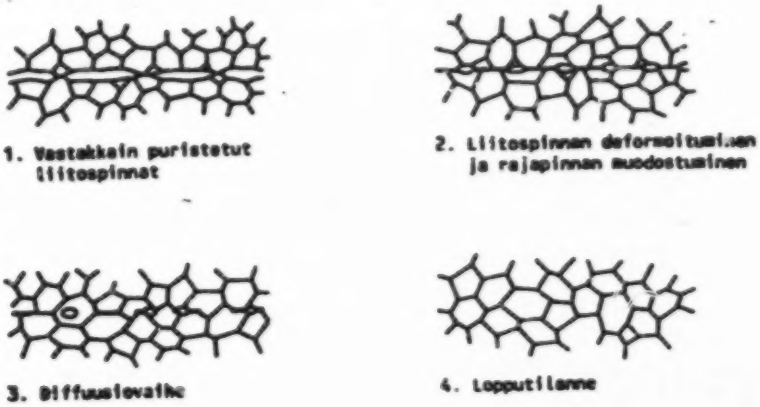

Figure 2. 1. Bond surfaces pressed against each other. 2. Bond surface deformation and alteration in boundary surface. 3. Diffusion phase. 4. Final arrangement.

Diffusion welding is generally understood as welding taking place in a solid state (so-called solid state diffusion bonding). However, diffusion welding is also used in a molten state (so-called liquid state diffusion bonding). In this variation either an additive plate, inserted between the joint surfaces, melts or constitutional changes occurring because of the diffusion lower the fusion temperature and cause

the bond area to fuse (Bartle, 1979). This form of diffusion welding is quite reminiscent of the hard solder process.

Output

The following workphases are generally associated with diffusion welding (Bartle, 1979; Bailey and Borbidge, 1980; Klomp, 1972):

--Carefully dressed bond surfaces (ground and polished, if necessary, R_a = 0.2 - 0.4 μm) and are cleaned of dirt and grease (by acetone or some other solvent). Success with diffusion welding also presupposes that bond surfaces are sufficiently parallel.

--In bonding special types of materials,(e.g., ceramic components), a thin plate is inserted between the bond surfaces, which promotes diffusion between the pieces and avoids the formation of undesirable aggregates in the bond area.

--The bonded pieces are put into a vacuum or protective gas chamber, in which the pieces are heated and the bond surfaces pressed against each other. The temperature is usually selected in the range of 0.4 - 0.9 T_m (T_m = fusion temperature). The pressed surfaces to be used are generally under 7 - 21 N/mm^2 to avoid distortion. However, the diffusion welding of certain ceramic classes assumes the use of surfaces under tangibly greater pressures (see Tables 3 and 4).

--The pieces are usually heated for 10-30 minutes as a function of the materials to be bonded, the temperature and the pressure exerted. The welding parameters leading to maximum bond strength are generally determined experimentally.

Pressure is usually applied hydraulically or magnetically. The isostatic gas surface or the surface of the piece itself can also be used expediently.

Pieces to be welded are heated by electrical resistance, heat radiation or inductive current, Figure 3. The induction heating of ceramic components assumes that the ceramic piece conducts electricity or is lined with metal. Otherwise the parts are heated only in the area of a metallic insertion plate. The heat distribution of the pieces to be joined can be removed and the overheating of the bond area

and melting can be avoided by means of a metallic heat conduction tube, Figure 3. The heat from the bond area can also be distributed by means of an induction coil design (Pabst and Moergenthaler, 1976; Elsner, 1975).

Table 3. Diffusion welding of ceramic-ceramic bonds; bond types, parameters and bond properties.

Bond type 1	Process[1] 2	Additive 3	Parameters[2] 4	Strength[3] 5
Al_2O_3/Al_2O_3 (sintered) 6	K	Al_2O_3 (high-pur.) 7	1650 °C, 60 min, 300 MPa	
	K	-	1675 - 1800 °C, 15 min, 10 MPa	8 tens. strength 200 MPa
	S	Cu_2O	1230 °C, 10 - 30 min, in air 10	9 cutting strength 150 MPa
	S	Ti-25Cr-21V	1550 - 1650 °C	
	S	49Ti-49Cu-2Be	1000 °C, 10 min, vacuum 11	
	S	72Ag-28Cu 65Ag-20Cu-15Ti 95Ag-4Cu2O V_2O_3, Al_2O_3, HfO_2, MgO, $50V_2O_3$-$50Fe_2O_3$		
	S	50Ti-50Cu	1025 °C, 1 min	9 cutting strength 145 MPa
SiC/SiC	S	Si-Ti, Ti	1200 °C, 2 min, 20 MPa	
	S	Al	1000 °C, 2 - 3 min, 3 MPa	12 melt-sintered 150 - 240 MPa
	K	SiC + C	1450 °C, 30 min, 1500 MPa	12 melt-sintered 330 MPa
	K	SiC + Al + B + C	1650 °C, 30 min, 30 MPa	12 melt-sintered 540 MPa
Si_3N_4/Si_3N_4 (sintered) 6	K	Si_3N_4 (high-pur.) 7	1800 °C, 60 min, 3000 MPa	
	K	ZrO_2, $ZrSiO_4$		
	S		1330 °C	12 melt-sintered approx. 250 MPa
Al_2O_3/Si_3N_4	K	Al_2O_3, AlN	1800 °C, 30 - 40 min, 2000 MPa	
ZrO_2/MgO	S	Cu_2O	1200 °C, 1 - 10 min	9 cutting strength 45 MPa

13 [1] K = klassische (classical) Diffusionsschweissung, S = neuere (new) Diffusionsschweissung
14 [2] Parameter: Schweisstemperatur (°C), Schweisszeit (min), Pressdruck (MPa, kPa)
15 [3] Festigkeit (strength)

Key: 1-bond type, 2-process, 3-additive, 4-parameters, 5-strength, 6-sintered, 7-heat pressure, 8-bending strength, 9-cutting strength, 10-in air, 11-in vacuum, 12-four-point bending, 13-diffusion welding of closed area, S = diffusion welding of melt area, 14-set-up: heating temperature (°C), heating time (min), pressure surface (MPa, kPa), 15-at room temperature.

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Table 4. Diffusion welding of ceramic-metal bond; bond types, parameters and bond properties.

Liaison-type 1	Wilhelmsen-key 2	Name-[1] ation 3	Parameter[2] 4	Lujan[3] 5
Al_2O_3/Ni	-	K	1140 °C, 2 h, 100 MPa	[illegible] 7,5 Nm
	-	K	1300 °C, 30 min, 170 MPa	nulpistostatv. 7 200 MPa
	Cu-O-comp. 6	S	1065 - 1083 °C, 15 min-clm. 10	hrinpistostatv. 7 30 MPa
Al_2O_3/Niobium 99%	96Ag-4CuO	S		
Al_2O_3/Fe	-	K	1035 °C, 24 h, 100 MPa	[illegible] 12 Nm
	-	K	1150 °C, 1,5 min, 31 MPa	nulpistostatv. 7 200 MPa
	-	K	1700 °C, 30 h, 2000 MPa	univ. Lujan 11 220 MPa
Al_2O_3/Au	-	K	1000 °C, 4 h, 100 MPa	[illegible] 7,9 Nm
Al_2O_3/Fe	-	K	1375 °C, 1,5 min, 71 MPa	nulpistostatv. 7 200 MPa
	-	K	1410 °C, 0,5 h, 200 - 3000 MPa	vonLujan 12 75 MPa
Al_2O_3/Invar	96Ag-4CuO	S		
Al_2O_3/Cu	-	K	1025 °C, 15 min, 130 MPa	nulpistostatv. 7 130 MPa
	90Cu-10Ti	S	1050 - 1200 °C, 30 min	Leitk. Lujan 13 max. 145 MPa
Al_2O_3/Al	-	K	607 °C, 1,5 min, 700 MPa	nulpistostatv. 7 95 MPa
Al_2O_3/Pb	-	K	290 °C, 2 min, 1000 MPa	nulpistostatv. 7 55 MPa
Al_2O_3/Ta	Ti-20Cr-2V	S		
Al_2O_3/Ti	69Ti-28Cu-3Be	S		
Al_2O_3/Zircaloy 2	69Ti-28Be-3Be	S		
ZrO_2/Fe	-	K	1300 °C, 30 min, 2000 MPa	
SnO_2/Cu	90Cu-10Ti	S		Leitk. Lujan 8 8,3 MPa
SiC/Ni, Ni-30Cr, Ni-30Al, Ni-30Cr-8Al	-	K		

1) K = binodale Lücke diffusionsbinden, S = eutektische Lücke diffusionsbinden
2) Parameter: Schweißtemperatur (°C), Haltezeit (h, min), Preßdruck (MPa, MPa)
3) Verbindungsart

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Table 4 continued

Liitos-tyyppi 1	Välikappale-levy 2	Menet-tely[1] 3	Parametrit[2] 4	Lujuus[3] 5
SiC/Si, Mo, Ta	-	K		
SiC/Al, Ti, Cu, Fe	-	K	1000 °C, 4 - 20 h	
SiC/Cu, Al	Cu Cu-35 Fe	S	860 - 880 °C	Peelin kerr 19 MPa
Si_3N_4/Fe	-	K	1410 °C, 20 min, 100 - 3000 MPa	vetolujuus 75 MPa
Si_3N_4/AISI 304	Al	S	670 °C, 2 - 60 min	vetolujuus 45 MPa
Si_3N_4/AISI 316, Hastelloy X, Nimonic 75, 70Ni-19Cr-5Al	-	K		
Si_3N_4/Ti, Zr, Mo, W, Ta	-	K		
MgO/Pt Pd, Ni	-	K	Pt: 1090 °C, 2 h, 100 kPa	vetolujuus 5,2 Mn
MgO/Ni		S	1100 °C, 30 min	leik., lujuus 29 MPa
MgO/Al, Mo, Ta, Al	-	K		
MgO/Mo, Ta	49Ti-49Cu-2Be	S	1000 °C, 5 min, tyhjössä	
MgO/Cu	90Cu-10Ti	S S	1025 °C, 1 min 1.480 °C, 20 min	leik., lujuus 70 MPa vetolujuus 58 MPa
BaO/Pt, Au, Ag, Ni, Fe	-	K	1100 °C, 17 h, 100 kPa	vetolujuus 9,7 Mn
BaO/Zr	95Zr-5Be	S	1050 °C, tyhjössä	
TiC/Fe, Ta, W	-	K		
AlN/Fe, Ta, W	-	K		
UO_2/Fe, Mo, Zr	Fe-49Ti-49Cr-2Be Mo-49Ti-49Ag-4Be Zr-95Zr-5Be	S		

1) K = kiinteän tilan diffuusiohitsaus, S = sulan tilan diffuusiohitsaus
2) Parametrit: kuumennuslämpötila (°C), kuumennusaika (h, min), puristuspaine (kPa, MPa)
3) Kuumennuslämpötilassa

Key: 1-bond type, 2-interface plate, 3-procedure, 4-parameters, 5-strength, 6-Peel's coefficient, 7-four-point bending, 8-alloy, 9-three-point bending, 10-air atmosphere, 11-bending strength, 12-tensile strength, 13-cutting strength, 14-diffusion welding of closed area, S = diffusion welding of melt area, 15-set-up: heating temperature (°C), heating time (min), pressure surface (MPa, kPa), 16-at room temperature, 17-in vacuum.

Types of Bonds and Parameters

Developmental work in diffusion welding technology has rendered the production of very many kinds of ceramic-ceramic and ceramic-metal bonds possible. The

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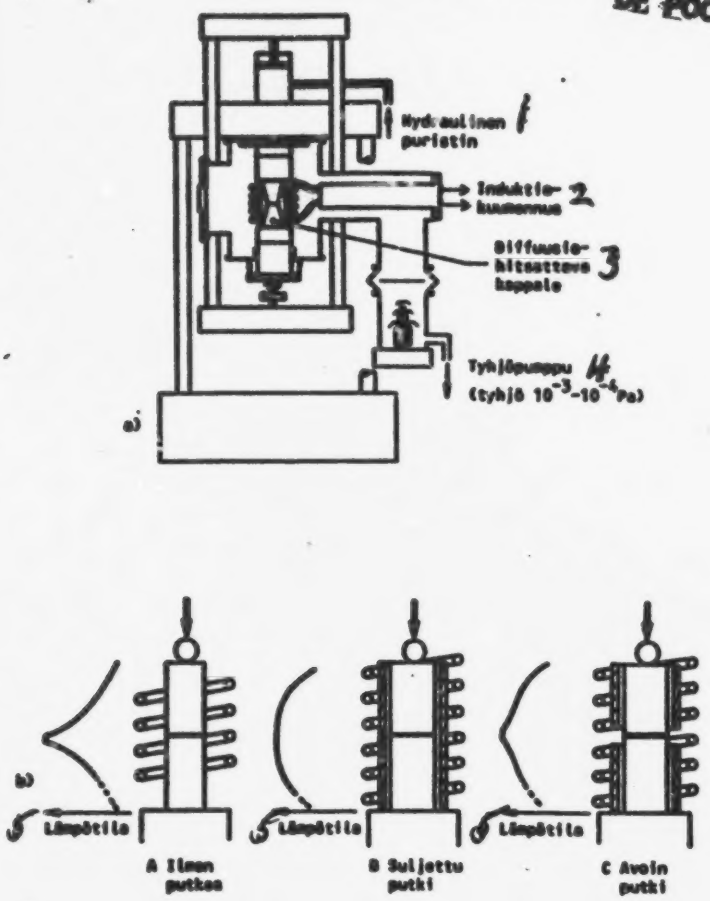

Figure 3. Diffusion welding brought about by induction heating of ceramics components (Pabst and Moengenthaler, 1976): a) apparatus, b) use of heating sets and effect of heat distribution (A, B, C). A-without tube, B-closed tube, C-open tube. Key: 1-hydraulic pressure, 2-induction heating, 3-piece for diffusion welding, 4-vacuum pump (vacuum 10^{-3} - 10^{-4} Pa), 5-temperature.

sensor structures shown in Figure 4 are examples of typical bond applications. The bonds have also been used well in the production of thermocouples, Figure 5. The structural solution in question has made it possible to raise the reaction rate of thermocouples as much as five times over the customary solution in Figure 5. This type of execution has tangibly added economy in power and in the fusion process.

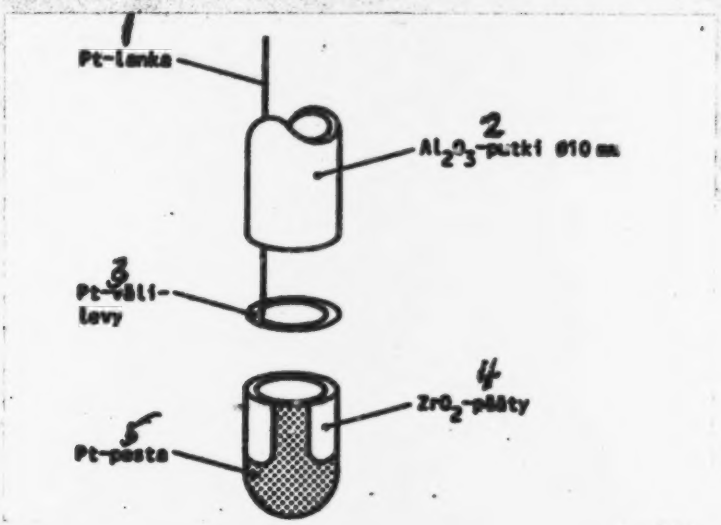

Figure 4. Diagram of O_2 analyzer, ZrO_2 gable piece, Pt-plate and Al_2O_3-tube are joined by diffusion welding. Temperature range 600-1400°C (Bailey and Borbidge, 1980). Key: 1-wire, 2-tube, 3-plate, 4-gable, 5-paste.

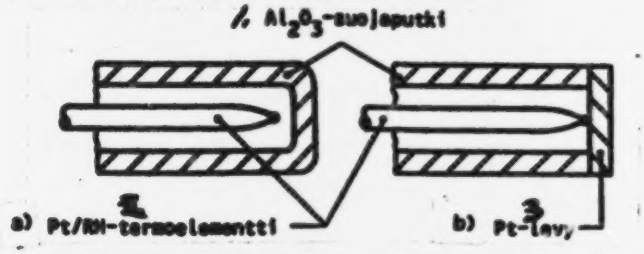

Figure 5. Good use of diffusion welding in rapidly reacting thermocouple production. In figure, a) traditional and b) diffusion welding construction (Bailey and Borbidge, 1980). Key: 1-protective tube, 2-thermocouple, 3-plate.

Tables 3 and 4 present a summary of special possibilities for diffusion welding covering Japanese plans (Associate Professor K. Nishimoto: written response to VTT inquiry of 14 May 1984). The diffusion welding parameters and the strength values of most types of bonds are shown in the Figures. Some of the general types of bond diffusion welding are treated in more detail by means of the following examples.

Bond Type Al_2O_3/Nb

Al_2O_3 diffusion welding makes use of many Nb interfaces whenever the application temperature rises to about 1400-1500°C, Figure 6. The heat expansion coefficients of Al_2O_3 and Nb, being quite close to one another (Table 1), give aggregates of Al_2O_3/Nb good resistance to thermal loads (Elssner and Petzow, 1973).

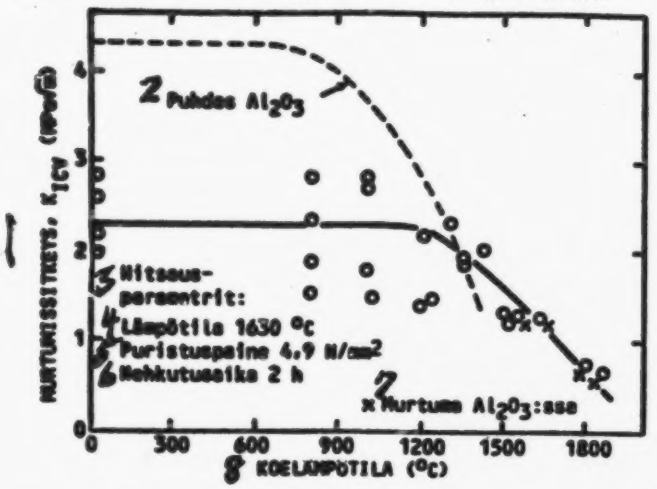

Figure 6. Breaking strength K_{ICY} of diffusion welded Al_2O_3/Nb bond and of Al_2O_3 (e.g., seam) at different temperatures (Moergenthaler et al., 1979). Key: 1-breaking strength, 2-pure, 3-welding parameters, 4-temperature, 5-pressure surface, 6-heating time, fracture, 8-test temperature.

Figure 7 shows that an aggregate with Al_2O_3/Nb diffusion welding can be successful in many different combinations of parameters. The increase in the heating temperature and the pressure area seem to raise the breaking strength of the bond very definitely. On the other hand, the effect of the heating period becomes evident only with long periods of heating ($\geq$ 8 hours).

Bond Type Al_2O_3/Pt and Al_2O_3/Au

Figure 8 presents the results of hot tensile tests of Al_2O_3/Pt and Al_2O_3/Au combinations at different temperatures. The heat resistance of the Al_2O_3/Pt pair

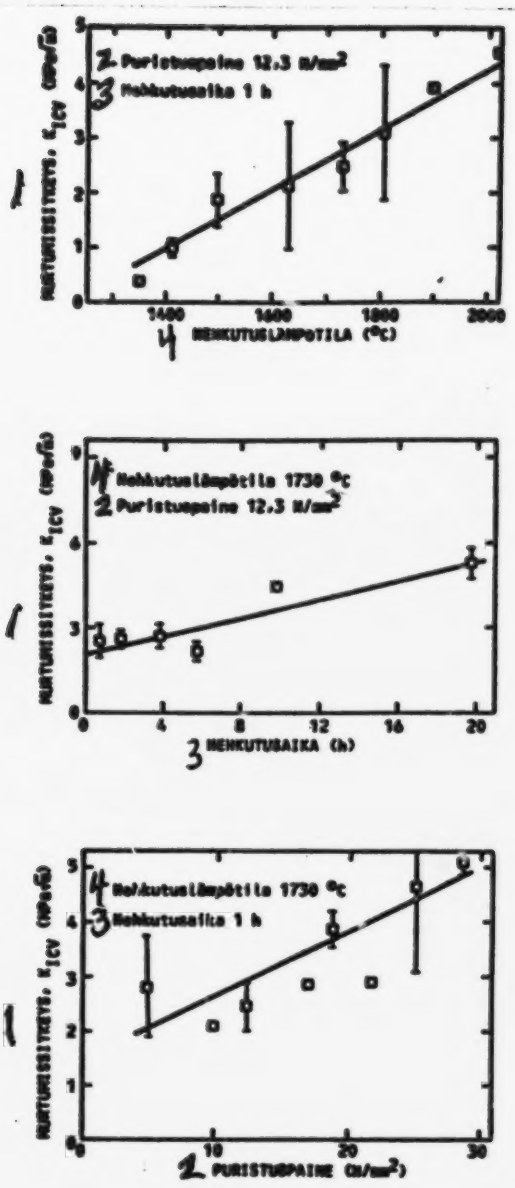

Figure 7. Breaking strength K_{ICV} of diffusion welded $Al_2O_3/Nb/Al_2O_3$ bond (e.g., seam) as a function of welding a) heating temperature, b) heating period and c) pressure surface. Key: 1-breaking strength, 2-pressure surface, 3-heating period, 4-heating temperature.

bond seems good at least to 2000°C. The heat expansion of the members of the aggregate, being close to each other (Table 1), contributes to the thermal stability of the structure.

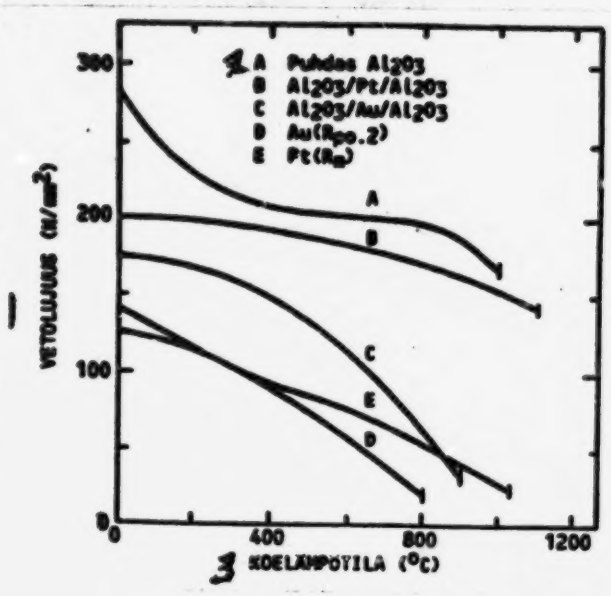

Figure 8. Tensile strength of $Al_2O_3/Pt/Al_2O_3$ and $Al_2O_3/Au/Al_2O_3$ bonds and corresponding basic materials at various test temperatures (Bailey and Borbidge, 1980). Key: 1-tensile strength, 2-pure, 3-test temperature.

The Al_2O_3/Au aggregate proves to have clearly worse heat resistant properties than the Al_2O_3/Pt aggregate (see Figure 8 and Table 1). Nevertheless the range of use of one Al_2O_3/Au bonded pair is, for example, the diffusion welding of composite structures (Bailey and Borbidge, 1980).

Bond Type Al_2O_3/Ni

Al_2O_3 diffusion welding is also successful with the help of an Ni interface plate (Table 4). However, under oxidising application conditions, the Al_2O_3/Ni at the boundary surface tends to change to a spinel-type Ni-oxide layer (type $M'O\ M''_2O_3$, where M' and M" are metals), where the increased thickness lowers the bonding strength. For this reason the Al_2O_3/Ni combination is used only in vacuum and protective gas applications, e.g., discharge bulbs. In addition the Al_2O_3/Ni bond pair can advantageously unite the Ni component, for example, to a structure made of stainless steel (Bailey and Borbidge, 1980).

Bond Type Si_3N_4/Zr

According to Table 3 a heat compressed Si_3N_4 plate can be used in diffusion welding with Si_3N_4 produced by sintering. However, bonding success presumes the use of very high pressure surfaces. The diffusion welding of Si_3N_4 pieces succeeds instead with noticeably lighter pressure surfaces when zirconium (Zr) is used as an interface plate. According to Figure 9, this combination can be diffusion welded only within certain welding parameters. The reaction kinetics of the Si_3N_4/Zr bonding pair determines the temperature below which bonding does not take place. The upper limit of the heating temperature, for its part, determines the brittleness of the oxide layer grown onto the Si_3N_4 surface, which can lead to fracture of the bond in the diffusion welding cooling stage. For example, from the bonding viewpoint a safe temperature range (150-200°C, Figure 9) is still a safe interval with an hour's heating.

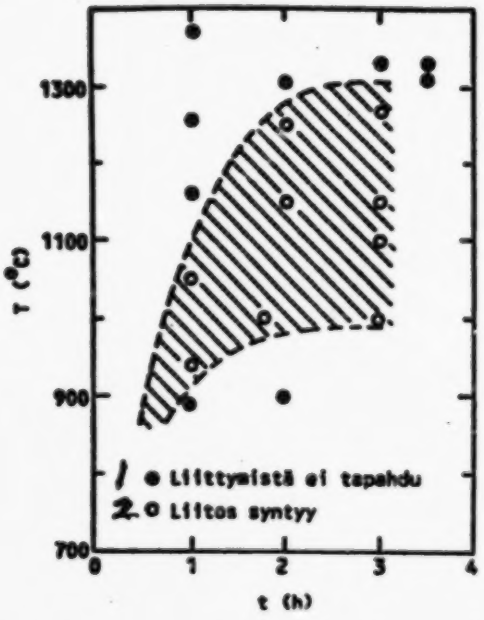

Figure 9. Effect of diffusion welding and its temperature on the formation of a hot-pressed bond between Si_3N_4 and Zr (pressure surface 10 N/mm^2) (Elsaner et al., 1980). Key: 1-no bonding occurs, 2-bond forms.

Glass-Metal Bonds

The diffusion bond between the superalloy Inconel-X 750 (73%, 15.5% Cr, 7% Fe, 2.5% Ti) and barium silicate glass is used nowadays in combustion engines and heat exchangers. They are produced by patented processes (Kelsey and Siegel, 1984), where a thin chromium oxide-nickel-titanium layer is preoxidised onto a superoxide surface. After this the combination is bonded in an air atmosphere (temperature 1095°C, time 1 minute, pressure 14 N/mm^2). In this process the surface of the barium silicate glass melts. The melt crystallises in the cooling stage into the same crystalline shape as an oxide layer produced by preoxidation on the superalloy surface. The generation of the same crystal during bonding secures a hard grip, noted for a permanently good thermal and mechanical load.

These processes make it possible to produce composite structures, among other things, in which the good anticorrosion and fireproof resistance of ceramics is united with the good strength and fatigue resistance of metals (Kelsey and Siegel, 1984.

2.2.2. Electron Beam and Laser Welding

The research on bonding technology for ceramic components has also been directed toward fusion welding processes. In connection with them experiments have been conducted with electron beam and laser welding, which can be considered applicable to these ends because of their low deformation level and good working precision.

The principle of electron beam welding is illustrated in Figure 10. The inertial energy of the electron beam directed at the workpiece changes to heat energy when it strikes the piece. Then the rapid heating and vaporization of the material assumes the shape of the seam in a brief span of time ($\sim$1-10 ms) and, if need be, to a good depth ($\sim$ 0.1-500 mm). Electron beam welding is usually conducted in a vacuum chamber ($\sim 10^{-1} - 10^{-3}$ Pa, Figure 10), because this leads to the best feasible results (Karppi and Vilpas, 1984).

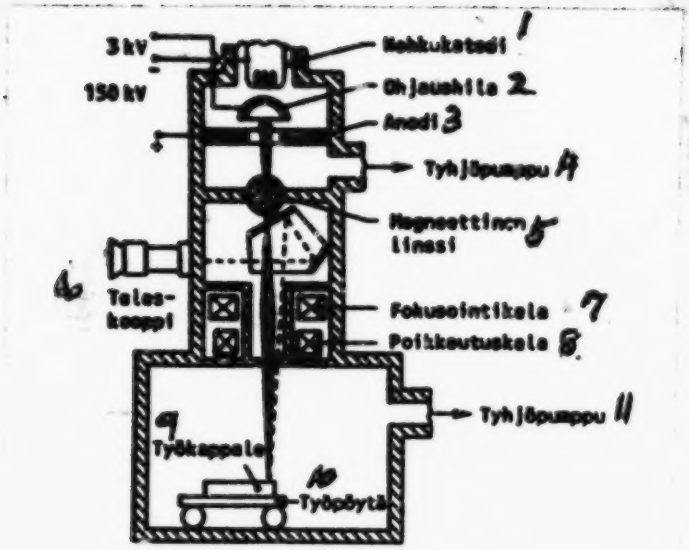

Figure 10. Apparatus and active principle of electron beam welding. Key: 1-heating coil, 2-control grid, 3-anode, 4-vacuum pump, 5-magnetic lens, 6-telescope, 7-focussing dial, 8-direction dial, 9-workpiece, 10-worktable, 11-vacuum pump.

The heat energy needed to form the laser welding seam is produced by the laser beam, Figure 11. Depending on the nature of the laser beam, the process is assigned to pulsed or continuous laser welding. High power creates continuous laser welding (CO_2-laser, $P \sim 3$-10 kW), which is very reminiscent of electron beam welding (so-called keyhole welding or piercing beam welding) in its seam-forming principle. The most noticeable advantage of the laser over the electron beam is the possibility of welding under atmospheric pressure (see Figures 10 and 11), at which time the size of the workpiece does not limit welding success. However, the use of laser welding is limited by the definitely higher acquisition costs over the electron beam welding equipment of a size suitable for welding ($P \geq 3$-5 kW). Therefore laser welding is generally used only in this plate technology (Neuronen, 1984).

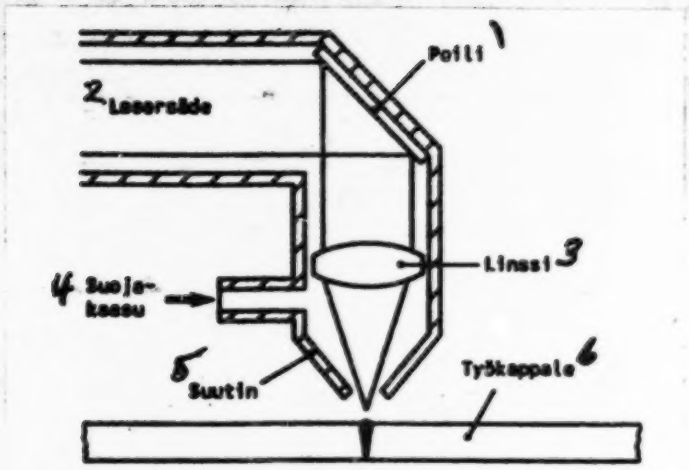

Figure 11. Active principle of laser welding. Key: 1-mirror, 2-laser beam, 3-lens, 4-protective gas, 5-jet, 6-workpiece.

Electron beam and laser welding can be used in bonding ceramic components by directing the beam to the metallic component or matellic part, which is soldered or diffusion-welded firmly to the ceramic surface. Bonding can also be achieved by fusion welding, by fusing the basic materials tightly to each other. Laser welding is considered particularly good for suitable fusion welding. Among others, Al_2O_3 joints are welded with laser beams, achieving a bonding strength nearly equal to the strength of the basic material. (IIW-Doc., 1980).

The use of electron beam welding in fusion welding assumes that the ceramic material is first made an electrical conductor by preheating, metallizing or graphite lining. This can eliminate the changing electrostatic field interfering with the electron beam in the bonded area. The electron beam welding of Al_2O_3 and metal pieces (W, Mo, Nb, Nb-0.5%Zr, Ta and Kovar) has been researched in laboratory tests. In these tests the ceramic component (Al_2O_3) was preheated to a temperature of 1150-1350°C. In the electron beam welding tests they used an exciting voltage of 90 kV

and a current of 2 mA (i.e., a 180 W beam effect. The welding transfer rate was changed 12-75 cm/min. For example, a tensile strength level of 40 N/mm^2 was reached in tests of bonding between Al_2O_3 and Ta (Gruenling, 1973).

Research on ceramic bonding technology is also directed toward the utilization of electron beam and laser welding. In these processes the electron beam and laser beam are used to bond ceramic and metallic components by diffusion or solder welding, or by fusion welding.

Research work associated with bonding technology for ceramic components is continuing intensively. There will probably be heavy use of diffusion welding. The utilization of electron beam and laser welding is also developing. The object of the developmental work is to make up for unsatisfactory and poor results by faster and more certain procedures for continuous welding processes.

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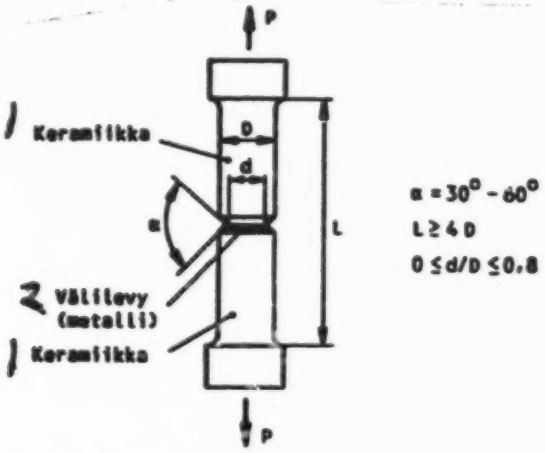
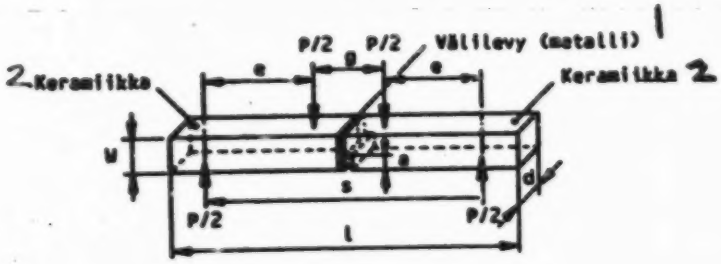


3 Murtumissitkeys (MPa/m):

$$K_{ICV} = \frac{P_c}{D^{3/2}} \left(1{,}72 \frac{D}{d} - 1{,}27\right)$$

Tensile Test. Key: 1-ceramic piece, 2-plate (metallic), 3-breaking strength.



$$\frac{g}{W} > 2,\ 1 \geq 4{,}2\ W$$

$$\frac{s}{W} \geq 4,\ 0 \leq \frac{a}{W} \leq 0{,}6$$

$$2 \leq \frac{W}{d} \leq 8\ \left(\frac{W}{d} = 2\right).$$

3 Murtumissitkeys (MPa/m):

$$K_{ICV} = \frac{3P_c \cdot e/\bar{a}_c}{dW^2} \left[1{,}99 - 2{,}47 \left(\frac{a}{W}\right) + 12{,}97 \left(\frac{a}{W}\right)^2 - 23{,}17 \left(\frac{a}{W}\right)^3 + 24{,}80 \left(\frac{a}{W}\right)^4\right]$$

Bending Test. Key: 1-plate (metallic), 2-ceramic piece, 3-breaking strength.

Published by Technical Research Centre of Finland
Vuorimiehenkatu 5
SF-02150 Espoo 15, Finland
phone internat. + 358 0 4561
telex 123572 vttlib sf

Name, number and report code of publication
VTT Research Notes 481 FI+VTTTIED-85/481

Date
August 1985

Project number
634609-8

Authors
Vilpas, Martti

Name of project
Korkeissa lämpötiloissa käytettävät...

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Title
JOINING OF CERAMICS FOR HIGH TEMPERATURE APPLICATIONS

Abstract
The report is a summary of literature survey to the methods for joining ceramics to ceramics or ceramics to metals for high temperature applications. Also mechanical properties and potential applications of the joints are considered.
The joining of ceramics is usually carried out by brazing or diffusion bonding. Especially the latter has been found useful and increased the applications of joined ceramics. Recently also the possibilities to use electron beam and laser beam welding for joining of ceramics have been investigated. The joining of ceramics has found numerous applications, typical for high operating temperatures being different sensors and thermocouples.

Activity unit
Metals Laboratory, Metallimiehenkuja 6, SF-02150 Espoo, Finland

ISSN and key name
0358-5085 Tiedotteita - Valtion teknillinen tutkimuskeskus

ISBN
951-38-2381-4

Language
Finnish, Engl. abstr.

Class (UDC)
621.792.4/.6:666.63

Key words
ceramics, joining, welding, heat resistant materials

Sold by
Government Printing Centre
P.O. Box 516
SF-00101 HELSINKI
phone internat. +358 0 538011

Pages
27 p.

Price
FIM 30

Note

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